Relationship between tunnel magnetoresistance and magnetic layer structure in EuO-based tunnel junctions investigated using polarized neutron reflectivity

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This paper presents a study of the depth-dependent magnetic structure of a EuO magnetic tunnel junction having a Gd electrode, Si/Cu/EuO/Gd/Al. Related samples that are patterned exhibit large tunneling magnetoresistance as high as 280%. Though Gd has a much higher coercivity than EuO in bulk, magnetometry reveals no "steps" in the hysteresis loop as expected for a true antiparallel alignment of the EuO and Gd layer magnetizations. Using polarized neutron reflectometry to measure the structural and field-dependent magnetic depth profile at 5 K, we determine that the Gd and EuO layers have similar coercivities and that the Gd layer exhibits an anomalously small magnetization at all fields. Polarized neutron reflectometry results also suggest that the chemical density of the Gd layer is not that of bulk Gd. The differences of the structural and magnetic behavior of the Gd layer relative to bulk may be the key in optimizing the tunnel magnetoresistance in these samples. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2837873]

As determined by Julliere,[1] the magnitude of the tunnel magnetoresistance (TMR) for a magnetic tunnel junction (MTJ) depends on the spin polarization (P) of the ferromagnetic electrodes, which has spurred the ongoing search for ferromagnets (FMs) with high spin polarization, ideally $P = 100\%$. Typical TMR devices involve tunneling polarized spins from a FM, through an insulating, nonmagnetic barrier, into a second FM. Alternatively, high TMR can be achieved by tunneling through a ferromagnetic tunnel barrier, which produces a highly polarized current via the spin-filter effect.[2,3] The spin-filter barrier in this approach is sandwiched between a FM and a nonmagnetic counterelectrode, and the TMR results from the relative magnetic alignment of the FM and the barrier. Such a quasi-MTJ structure is investigated in this study, with ferromagnetic EuO as the tunnel barrier, which is known to be an efficient spin filter.[4] In a recent study, Negusse[5] showed the effect of electrode material on the chemical nature of the EuO/electrode interface. Three important factors affecting the chemical and magnetic properties, and thus the spin-filtering efficiency, of a EuO barrier are (1) formation of Eu_2O_3, a stable nonmagnetic oxide, which is undesired, (2) the structural properties of the electrodes used for injecting and detecting the current, and (3) the quality of interfaces which is crucially important in spin-conserved transport. Despite these challenges, large TMR of up to 280% was observed in patterned Cu/EuO/Gd quasi-MTJs.[6]

To explore issues with optimization of the TMR in these structures, we characterized a quasitunnel junction comprised of a Si/1 nm Cr/5 nm Cu/7.5 nm EuO/12 nm Gd/14 nm Al layer structure using superconducting quantum interference device magnetometry (SQUID) and polarized neutron reflectometry (PNR). This sample was grown using thermal reactive evaporation and is identical in preparation to the patterned junction which exhibited large TMR[6] though the planar area was significantly larger (1×1 cm^2). Details of the growth are provided elsewhere.[4]

Figure 1 shows the field-dependent magnetic moment at 5 K, as measured using SQUID magnetometry. The hysteresis loop exhibits a sharp field transition near 0.015 T followed by a gradual approach to saturation beginning near 0.02 T. The shape of the hysteresis loop, thus, suggests that the EuO and Gd layers are coupled to some extent. Specifically, no obvious plateau is observed between the negative and positive saturation states as would be expected for true

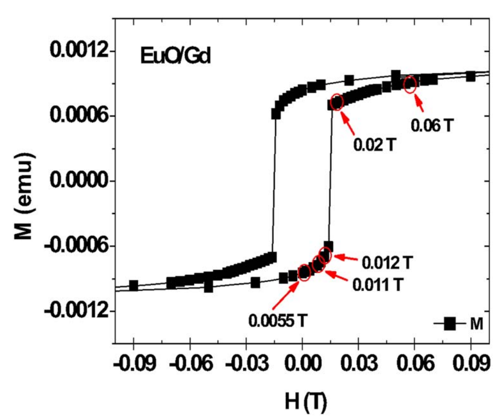

FIG. 1. (Color online) Magnetic moment of magnetic tunnel junctions with Gd electrode taken at 5 K using SQUID. Circles represent fields where a PNR measurement was obtained.

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antiparallel alignment of the magnetizations of the EuO and Gd layers. The absence of antiparallel alignment is surprising, since bulk EuO and Gd have very different low temperature coercivities. This point is critical, as independent alignment of the EuO and Gd layers is assumed to be a necessary condition for maximizing the TMR. We note that these hysteresis data are typical of the patterned quasitunnel junctions that exhibit large TMR.[6]

We used PNR to determine the depth-dependent magnetization of the sample and to identify the magnetic structures of the EuO and Gd layers associated with the features in Fig. 1. To reproduce the conditions for the magnetization measurements (Fig. 1), PNR data were obtained at a series of fields after field cooling from room temperature to 5 K in −0.7 T. For the PNR experiments, neutrons were polarized parallel to the applied field in the sample plane, as described in Ref. 7. The reflectivity data were corrected for instrument background, polarizing element efficiencies (typically >97%), and beam footprint. There are four reflectivity cross sections: R^{++} and R^{--} labeled as nonspin flip (NSF) as the neutron retains its original polarization, and R^{+-} and R^{-+}, labeled as SF, where the neutron spin rotates 180°. The chemical scattering length density (ρ_{chem}), the nuclear scattering ability of the material, can be inferred from fits to the reflectivity data.[7–9] In addition, the splitting between the NSF reflectivies is sensitive to the component of the magnetization aligned along the field axis and provides information concerning the chemical composition of the film. SF reflectivity is sensitive only to that magnetization component perpendicular to the applied field. To obtain the chemical and magnetic profile as a function of depth, the NSF PNR data were fit[10] with the REFLPAK (Ref. 11) and GAREFL (Ref. 12) software suites. While some SF scattering was detected, the data showed no obvious field dependence and appeared to be an artifact only. Thus, the fits reveal only the component of the magnetization parallel to the applied field.

After field cooling, a measurement was taken at 0.7 T during the initial field cycle in order to obtain information at saturation. The reflectivity data have a similar appearance to those data shown in Fig. 3, though the splitting between the R^{++} and R^{--} cross sections is somewhat more pronounced. Structurally, fits to the data reveal the Gd layer to be 7 nm thicker than the expected value (12 nm) and the ρ_{chem} exhibits a 65% reduction in value from the nominal value of 1.97×10^{-6} Å^{-2}. The possibility of layer impurity is further supported by the large roughness present at both the EuO/Gd interface (~6.3 nm) and the Gd/Al interface (~4.2 nm), indicating smearing or alloying, illustrated in Fig. 2.[13] Fits to the 0.7 T reflectivity data also reveal a EuO layer magnetization corresponding to a moment per Eu 1.16 times higher than the expected $7.0\mu_B$ and a similar increase in the ρ_{chem}, indicating possible layer strain. However, the Gd magnetization corresponds to a moment per Gd of less than 10% of the bulk value of $7.55\mu_B$. The reduced magnetization of the Gd layer suggests that it is not behaving similar to the bulk even at 0.7 T and could imply an impure Gd layer due to oxidation, alloying, or the formation of domains.

After field cycling to −0.7 T, a positive field was applied in the sample plane. Figure 3 shows the evolution of the PNR data at low field during the second field sweep. Measurements were taken at 0.0055 T [Fig. 3(a)], 0.011 T [Figs. 3(b) and 3(c)], 0.012 T [Fig. 3(d)], 0.02, and 0.06 T with two successive measurements taken at 0.011 T to attempt to capture the moment relaxation. Noticeably, the EuO field reversal does not occur sharply at 0.014 T as expected from the magnetization data but takes place near 0.011 T. The primary indication of this transition is evident in the PNR data just below $Q=0.02$ Å^{-1}, where the gap between the two NSF cross sections collapses [Fig. 3(c)]. As the EuO layer completes its transition, R^{++} and R^{--} are reversed [i.e., Fig. 3(d) is similar but opposite to Fig. 3(a)], and the gap between these cross sections reemerges.

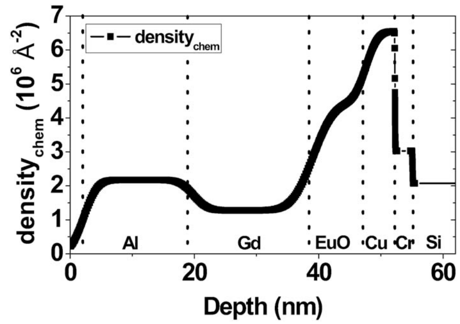

FIG. 2. Chemical profile as a function of depth illustrates large roughness at the EuO/Gd interface determined from fits to the reflectivity data.

The net magnetization of the combined Gd and EuO layers was determined from the PNR data in Fig. 3 and it virtually matches that obtained from bulk magnetometry at all fields, as demonstrated in the inset of Fig. 4. Our most significant result, however, is the field variation of the magnetization of the *individual* EuO and Gd layers that was also extracted from the PNR fits (top and bottom of Fig. 4, respectively). Focusing first on the EuO magnetization, the fits

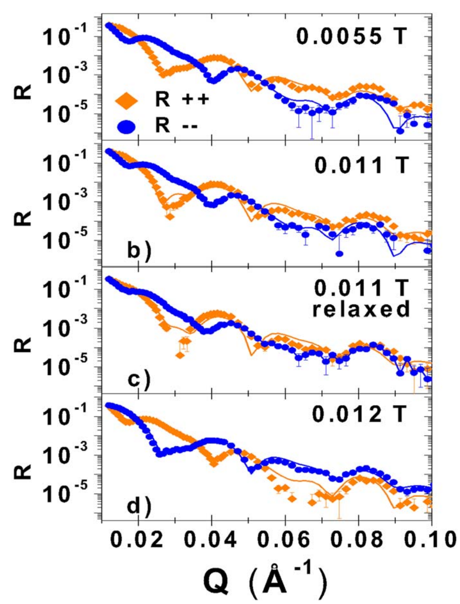

FIG. 3. (Color online) (a) PNR reflectivity data taken in a field of 0.0055 T. (b) PNR reflectivity data in a field of 0.011 T. (c) PNR reflectivity data repeated at 0.011 T. (d) PNR reflectivity data in a field of 0.012 T. All data are taken at 5 K. Lines correspond to fits to the data.

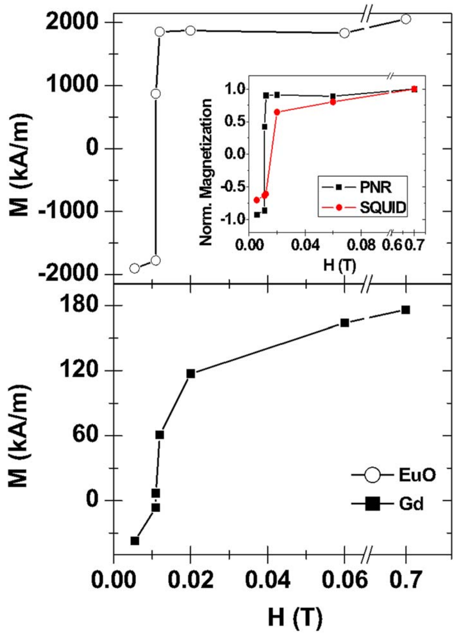

FIG. 4. (Color online) Comparison of the individual magnetization of EuO and Gd layers as a function of applied field as determined from fits to the PNR data. Bulk values for EuO and Gd are 1867 and 2169 kA/m, respectively. Inset compares SQUID and PNR measured magnetizations normalized to 0.7 T. Lines are guide for the eyes.

indicate that at 0.0055 T, the EuO magnetization is opposite and nearly equal (Fig. 4) to that obtained at 0.7 T. The EuO layer has a high remanent magnetization and has not yet reversed parallel to the field. Increasing the field to 0.011 T, the EuO magnetization decreases slightly to 86% of its saturation value, but it remains antiparallel to the applied field. Immediately repeating the measurement at 0.011 T, the reflectivity data captured the time evolution of the EuO moment relaxation. The drop in the EuO moment (Fig. 4), to $\sim\frac{1}{2}$ of the value at 0.7 T, and the collapse of the gap between the R^{++} and R^{--} data indicate that the EuO is realigning parallel to the field, possibly via the formation of small domains across the sample plane. The measurement taken at 0.012 T supports this assertion as the R^{++} and R^{--} have switched and the EuO magnetic SLD, which is directly proportional to the magnetization of the material, has increased to 90% of the 0.7 T value. The EuO magnetization remains near 90% of the saturation value in 0.02 and 0.06 T. The bulk of the EuO magnetization is, thus, essentially recovered by 0.06 T.

The field-dependent behavior of the Gd magnetization, shown in the bottom of Fig. 4, contrasts sharply with that described above. Similar to the EuO magnetization, the Gd magnetization at 0.0055 T is also antiparallel to the field, but it is reduced to ~20% of the value observed at 0.7 T. In a field of 0.011 T, the Gd layer magnetization has decreased by another factor of 10 to $0.18\mu_B$ and is nearly 0. We speculate that the layer is entirely broken up into small (<100 microns), in-plane domains. Along with the EuO, the Gd magnetization reverses direction near 0.014 T, but the Gd net magnetization remains small. The Gd magnetization increases very gradually as the field is increased to 0.7 T, suggesting that the Gd moments within the in-plane domains align very slowly. Overall, the Gd magnetic SLD is consistently smaller than its bulk value at all fields, and its field-dependent behavior is clearly tied to that of the EuO layer magnetization.

In conclusion, our PNR results indicate that the field-dependent EuO and Gd magnetizations are correlated and that antiparallel alignment of these layers is never achieved. There is a sharp EuO transition near 0.011 T followed by gradual realignment of in-plane domains within the Gd layer beginning near 0.02 T. In addition, the Gd magnetization is significantly less than that corresponding to the bulk moment per Gd of $7.55\mu_B$, even in high fields. While we have considered only a simple model in which the reduced magnetization is uniform throughout the entire Gd layer, it is also possible that this magnetization reduction is higher, but localized to only a small (approximately nanometer) region near the interface. The roughness of the structural interface suggests that the EuO and Gd layers may not entirely be chemically distinct. Fits to the data indicate that there may be an oxide formation, such as Gd_2O_3, or alloying of the Gd layer with adjacent layers. These structural issues may inhibit the anticipated antiparallel alignment of the EuO and Gd layer magnetizations. Instead, it is possible that the large TMR observed in related, patterned tunnel junctions[6] may originate from independent switching of in-plane domains within a layer that is not purely Gd. Without further optimization of the interfaces and magnetic behavior, it is difficult to determine if the formation of these domains is the cause of the high TMR measured or the hindrance to greater consistency in these types of tunnel junctions.

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[13] We note that the roughness observed at the EuO/Gd interface is consistent with that observed in one similar sample and three others with different EuO and Gd layer thicknesses.